FOREIGN DEVELOPMENTS IN COAL GASIFICATION

by Harry Perry
Director of Coal Research, Bureau of Mines
U. S. Department of the Interior

INTRODUCTION

The manufacture of either producer, synthesis, or higher heating value gases from coal continues to attract worldwide attention even though large new reserves of natural gas have been discovered and production of gas from oil rather than coal appears to be more economic in many countries at present. Because proved reserves of coal are much larger than those of oil and gas, coal is expected ultimately to be the raw material used to supplement these more convenient energy forms. The timing, however, will depend on how rapidly new reserves of oil and gas are discovered, their price, and also on the success of research and development efforts on coal conversion processes.

Many factors influence the degree of interest in gasification research. For example, a review of the Soviet literature shows a sharp decline in research on coal gasification starting about 1959 and indicates the shift in emphasis to other fuels. In many countries, the reduction in the number of miners employed as a result of a decline in total coal production or because of greater productivity per man has caused serious economic dislocations and social problems. For this reason and because of a desire to use indigenous resources rather than imports, there is still a good deal of interest in the development of new processes for economically converting coal to either oil or gas. Either of these potential outlets promises large new markets for coal.

Conversion processes that make gas from coal can be classified best by the type of gas that is produced. Producer gas, enriched producer gas, water gas, enriched water gas, synthesis gas, and synthetic methane are the major types of products that are usually made. Processes are operated at either atmospheric or superatmospheric pressures. Coal, coke, and char can be gasified in fixed beds in fluidized beds, and by entrainment.

In the U.S. after World War II, interest in gasification was centered upon the production of synthesis gas for subsequent use in the manufacture of either liquid or gaseous fuels. Since most synthetic fuel processes use gas at elevated pressures, significant savings are possible if pressure gasification is used. Moreover, because the manufactured products would be needed first in the densely populated eastern portion of the U.S., processes were sought that could use the strongly coking coals that are found in the east.

Interest in producer gas and water gas declined rapidly as natural gas became available in those markets at relatively low cost. However, as gas prices have continued to rise steadily during the past 15 years, there has been a revival of interest in the manufacture of a clean hot producer gas from coal for industrial use. In the more distant future, supplementing natural gas reserves with methane made from coal appears to be most promising.

In most foreign countries gas of lower heating value than methane is distributed, and processes making such gases have been extensively investigated. Lurgi generators, which operate at high pressure and produce a considerable percentage of methane in the synthesis gas, are particularly well suited for this type of market. However, in many foreign countries noncoking or weakly coking coals, which the Lurgi generators require, are usually readily available.

GASIFICATION WITH AIR AND STEAM

While the U.S. coal industry has indicated renewed interest[1] in the development

1/ Garvey, James R. Report in Proceedings of National Coal Assoc. 44th Anniversary Meeting, Washington, D.C., June 6-8, 1961, pp. 59-68.

of lower cost processes for the manufacture of producer gas for industrial use, as yet no new research has been initiated. This same situation exists in many foreign countries. In Germany, the vortex producer[2] developed by Ruhrgas is not

2/ Nistler, F. The Ruhrgas Vortex Coal Dust Producer. Coke and Gas, Vol. 19, No. 213, February 1957, pp. 54-57.

being operated, although a similar gas generator has been constructed in England at the Manvers Main Coke Works. At such installations, the manufactured producer gas is used to heat coke ovens, thus releasing the higher heating value coke-oven gas for distribution.

In Japan, research has been carried out at the Tokyo Institute of Technology[3]

3/ Kensuki Kawashimo, and Kozo Katayama. Combustion of Coal by Gasification. Bulletin Japanese Soc. of Mech. Eng., Vol. 4, November 1961.

on developing novel methods of making producer gas to be used for combustion in boilers, gas turbines, or industrial furnaces. In early experiments, 12 nozzles were used to inject two-thirds of the air tangentially into a fixed-bed producer. The other one-third was introduced beneath the rotating grate to reduce the carbon content of the ash. These tests proved that high-ash low-quality coals could be used to manufacture a satisfactory producer gas.

In more recent experiments, the producer was enlarged from 800 to 1,350 mm. in diameter. In July 1963, a rectangular producer 1,400 by 2,800 mm. with a number of cylindrical, horizontal, rotating grates was placed in operation and is now under test.

At the Industrial Research Institute of Hokkaido,[4] experiments have been

4/ Private correspondence, Rinzo Midorikawa, Director of Ind. Research, Institute of Hokkaido.

conducted on making producer gas from high-ash coals, lignite, and peat in a fixed bed. Coals with ash contents as high as 40 percent gave low thermal efficiencies, but lignites and peats were more easily converted and gave a gas of higher quality. Gas obtained from the drainage of methane from coal seams and containing up to 45 percent of air was converted to an oxygen-free gas in the producer, but the methane in the gas was decomposed in the process.

Although there has been a decline in interest in coal gasification in France because of the discovery of large deposits of natural gas, work on a fluidized-bed process was pursued until recently.[5/] Most of the tests used either lignite or

5/ Jequier, L., L. Longchambon, and G. Van De Putte. The Gasification of Coal Fines. J. Inst. Fuel, Vol. 33, No. 239, December 1960, pp. 584-591.

anthracite as fuel, although a few tests were made with bituminous coal. Temperatures had to be controlled closely to permit proper removal of the ash, but gasification of fines using a gasifier with specially designed bottom sections and carefully controlled injection of gases made this possible. As a result of this work, it was concluded that a large-scale plant could be built and operated successfully.

In Germany, increased capacity of water gas sets has been attained, and gases suitable for town gas distribution can be produced through oil carburetion during the down run period. This development will permit producers using coal and coke to compete effectively in some areas with gas produced from petroleum products alone.[6/]

6/ Domann, Von Friedrich. Methods of Gas Production - The Effect of New Types of Gas, Reprinted from Internationale Zeitschrift fur Gaswarme (International Gas Heat Journal, Vol. 12, 1963, pp. 177-179.

In another modification of the water gas process, the generation of water gas from coal rather than coke has been developed in order to be able to use indigenous noncoking coals and avoid the use of more expensive coke.[7/] In this process, the

7/ Domann, Von Friedrich and Hubert Schmitt. The Production of City Gas by Low-Temperature Carbonization of Coal in Tokyo andFreiburg (Breisgau)., Reprint from "Erdol und Kohl" Volume 12, 1959, pp. 883-90.

vessel above the gas generator is designed so that there is sufficient residence time for the coal to be carbonized before it reaches the gas generator.

GASIFICATION WITH OXYGEN-ENRICHED AIR AND STEAM

Starting in the mid-1950's, research in The Netherlands using oxygen-enriched air (50 percent oxygen) in fixed-bed gasifiers showed that such equipment could be operated using coke as the fuel and with the removal of ash as slag. These tests were continued until 1960, using oxygen concentrations as high as 90 percent because gases produced in this manner gave greater flexibility and were more economically attractive than either straight producer or water gas. Work on this process was discontinued with the recent discovery of large reserves of natural gas in The Netherlands.

The "bottom blown" gasifier requires only a relatively shallow fuel bed because of the more uniform distribution of the gasifying medium and the bottom flow characteristics of the fuel bed. However, the actual depth of bed required will depend on the rate of carbonization of the coal under the conditions in the gasifier.

Experimental work is still required to determine (1) the extent of the carry-over of fines and whether they can be recycled, and (2) the carbonization time of the coal in the gasifier. Large-scale experimental work under pressure is also needed to determine the heat losses in the water-cooled grates and the reduction in coal gas efficiency because of higher exit gas temperatures in the shallow fuel bed.

Badische Anilin and Soda Fabrik of Germany has developed a slagging gas producer operating at atmospheric pressure that can produce synthesis gas, using as a fuel either coke or a mixture of coke and hydrocarbons.10/ The design

10/ Duftschmidt, F. and F. Markert. Large-Scale Slagging Producers of Gas for Chemical Syntheses (Entwicklung von grosstechnischen Abstichgeneratoren zur Synthesegas-Erzeugung) Chemie-Ing-Techn., Vol. 32, No. 12, 1960, pp. 806-811.

of the slagging section was reported to be particularly satisfactory, and little difficulty has been reported from failure of refractories or with removal of the slag. A bath of slag is maintained at the base of the generator and slag is tapped intermittently. Satisfactory removal of the ash requires the addition of a limestone flux. The reaction of limestone with the coke ash to give a free flowing slag takes place mainly in the slag bath so that good distribution of the limestone and adequate residence time of the slag bath are required for satisfactory operation. The carryover dust is recirculated and is injected into the gasifier in a stream of the gasification medium. Forty to fifty percent gasification of dust is accomplished in one reinjection. Table 1 shows typical results obtained in this gasifier.

At the Regional Research Laboratory at Hyderabad, India, interest in gasification is centered about the use of coals containing from 25 to 35 percent ash. A slagging fixed-bed gasifier with a coal rate of 30 to 40 pounds per hour has been erected with a shaft 6 inches in I.D. and a slagging section 12 to 14 inches in I.D.11/ Four water-cooled tuyeres of 3/8 inch I.D. and 1/2 inch

11/ Some Aspects in the Design of Gasifier. Indian Chemical Engineer, July 1963.

diameter centrally located taphole have been provided. To date only preliminary experiments have been made using low-temperature coke as fuel and blast-furnace slag as a fluxing medium. These first experiments are being directed toward developing a satisfactory method of slag removal, and with a weight ratio of coke to slag of 7 to 3, satisfactory operations of from 45 minutes to 3 hours have been achieved.

Table 1.-Atmospheric pressure fixed-bed slagging gasification using coke

Material requirements1/	
Coke (87% C), pounds	27.8
Steam, pounds	18.9
Oxygen (90%), cu. ft.	240
Steam generated, pounds	10.8
Superheated steam, pounds	3.8
Gas analysis, volume percent	
CO_2	6
CO	62
H_2	31
N_2	1

1/ Per 1,000 cu. ft. CO + H_2

The Koppers Totzek process, which uses a wide range of solid and liquid fuels as feedstock, has been installed in a number of locations throughout the world.[12] However, no recent developments have been reported in oxygen gasification

12/ Osthaus, K. H., and T. W. Austen. Production of Gas from a Wide Range of Solid and Liquid Feedstocks by the Koppers Totzek Process. Gas World, Vol. 157, No. 4091, Jan. 12, 1963, pp. 98-103.

using entrained processes.

Improvements have also been reported[13] in the fluid-bed Winkler generator,

13/ Flesch, Von Wilhelm, and Gunter Velling. The Gasification of Coal in the Winkler-Generator. Erdol und Kohle, Vol. 15, No. 9, September 1962, pp. 710-713.

which is capable of gasifying both lignitic and bituminous coals. Modifications have been made in design to accommodate coking coals, and improvements in the generator are reported to give increased economies. By feeding a portion of the oxygen and steam into the upper part of the fluidized bed, it is possible to get high carbon conversions without slag formation.

Pressure Gasification

For many uses, synthesis gas is required at pressures of 30 atmospheres or more. Thus there has been a continuing interest in gasification processes operating at elevated pressures, since considerable savings are possible if the product gas is to be used at these pressures.

Because of limitations inherent in entrained and fluid-bed processes, the most economic method for producing synthesis gas from solid fuels at pressure appears to be fixed-bed operation removing the ash either dry or as a slag. Fixed-bed processes, however, require a sized fuel, and if coal is used, it must be noncoking or weakly coking. Dry ash removal requires an excess of steam and low throughput per unit volume of gasifier to keep the ash in a condition to be successfully handled. The removal of slag under pressure, however, introduces many difficult operating and design problems for which solutions are still being sought.

A number of Lurgi installations using dry ash removal have been installed in many countries. Operating results of the Westfield Lurgi Plant in Scotland, which is the most modern plant for which data are available, have recently been published[14] and are shown in Table 2.

14/ Ricketts, T. S. The Operation of the Westfield Lurgi Plant and High-Pressure Grid System. The Institution of Gas Engineers, Copyright Publication 633, May 1963, 21 pp.

A major improvement in Lurgi plant design has been accomplished recently in Germany by changing from water-cooled to steam-cooled grates, using the wet steam from the gasifier for this purpose, and by reducing the amount of steam to the gasifier. At Dorsten, the steam requirement was cut nearly in half, and the volume of gas produced could then by increased by 70 percent with only a 45-percent increase in oxygen consumption.

Table 2.-Operating results of Lurgi gasifier at Westfield

Material requirements 1/	
Coal, pounds	55.5
Steam, pounds	56.1
Oxygen, cu. ft.	238
Gas analysis, (crude), volume percent	
CO_2	24.6
C_nH_m	1.1
CO	24.6
H_2	39.8
CH_4	8.7
N_2	1.2

1/ Per 1,000 cu. ft. $CO + H_2$

Two other improvements in the Lurgi process have been introduced at Dorsten. In the first, the carbon monoxide in the crude gas is converted over a cobalt-molybdenum catalyst (by water gas shift) to carbon dioxide before the gas is purified. This results in a lower cost gas since no additional steam must be added as must be done in conventional carbon monoxide conversion where the excess steam is first removed from the raw gas and then added back before shift conversion. In the second improvement, air was added to the gasifying medium (this is satisfactory for certain types of gases in town use) and resulted in further improvement in plant operation. Steam decomposition was increased, thus reducing steam requirements, and because part of the oxygen was introduced as air, there was a substantial reduction in power requirements to produce oxygen.

In Germany experiments were conducted on pretreating the coal in the lock hopper of the coal feeding device to reduce the coking characteristics of the coals so that a wider range of coals could be used. The coal was treated with a mixture of carbon dioxide, nitrogen, and 1 percent O_2 at 200° to 250° C for 15 minutes; however, on the coals tested, little difference was noted in the operating results. These tests, along with others, indicated that for a given coal, coking problems were more severe in the smaller test generators than they are in the full-scale units. Because of the potential advantages of slagging operation, the most recent fixed-bed pressure gasification experimentation has used this type of ash removal. Research is underway on this method in the U.S., England, the U.S.S.R., and Germany.

The only recent report on gasification from the Soviet Union describes tests with a fixed-bed slagging gasifier operating at 5 atmospheres using anthracite as the fuel. These tests also involved refiring carryover dust and using up to 50 percent pulverized fuel.

In England, research on pressure gasification using a slagging fixed-bed gasifier has been conducted by the Gas Council at the Midlands Research Station and by the Ministry of Power at the B.C.U.R.A. Research Station at Leatherhead. This latter program was terminated in March 1962 after demonstrating that slagging operation under pressure could be accomplished. In addition, these experiments indicated probable methods for "scaling up" through the use of multiple tuyeres and has shown the potentials for using coal rather than coke as a fuel.[15]

15/ Masterman, S. O., and W. A. Peet. The Development of a Pressurized Slagging Fixed Bed Gasifier. Proc. of the Joint Conf. on Gasification (Institution of Gas Engineers and Institute of Fuel), Hastings, September 1962.

At the Midland Research Station, work is continuing on the slagging gasifier in a new pilot plant that was installed in 1962. The gasifier has a 3-foot I.D., operates at pressures up to 375 psi, and is designed to produce 5 million cubic feet per day of gas.

Until now the program has been aimed at the development of the hearth, slag tap, and slag discharge system. To avoid unnecessary experimental difficulties, graded coke has been used as a fuel, and the plant has been operated for as long as 90 hours at pressures of 300 psi and at 3 million cubic feet per day throughput. Slag is tapped intermittently and is quenched in water at the operating pressure. Experiments on the use of coal as a fuel were started in late November.

HYDROGASIFICATION OF COAL

Interest continues on the hydrogasification of coal, which was started at the Midland Research Station, but experimental work has been delayed because of the present emphasis in England on the use of petroleum feedstocks. For the type of gas distributed in England, the process selected[16] for study

16/ Dent, F. J. Chemical Engineering in the Production of Town Gas by Pressure Gasification Processes. Proc. of the Joint Conf. on Gasification (Institution of Gas Engineers and Institute of Fuel), Hastings, September 1962.

involves a two-stage hydrogenation of coal in fluidized beds at 70 atmospheres pressure. The first stage will operate at 1,475° to 1,560° F. to hydrogenate the volatile matter at lower temperatures, which thermodynamically favor a higher heat content of the gas. Char will be recycled to enable use of all types of coals. The second stage will operate at 1,650° to 1,740° F. to take advantage of the faster reaction rates at the higher temperatures required for the less reactive char. The residue of the hydrogenation is gasified, using oxygen and steam in a fluidized bed. The residue from the gasifier is used for steam production.

In similar research in Australia, tests have been made on the hydrogenation of brown coal in fluidized beds at pressures of 300 to 600 psi and at temperatures between 930° and 1,740° F. Early tests using cocurrent flow have been reported,[17]

17/ Birch, T. J., K. R. Hall, and R. W. Urie. Gasification of Brown Coal with Hydrogen in a Continuous Fluidized-Bed Reactor. J. Inst. Fuel, Vol. 33, No. 236, September 1960, pp. 422-435.

but the latest design uses countercurrent flow of reactants. The first experiments indicated that the reaction occurs in two stages with the first stage involving a rapid reaction of the oxygen-containing functional groups. The second stage involves a much slower reaction of the hydrogen with condensed carbon and required a much higher concentration of hydrogen and higher temperatures to get reasonable reaction rates.

Experiments with the countercurrent apparatus are still underway. To take advantage of the rapid hydrogenation at low temperatures during the initial period and yet obtain the higher temperatures required for the second stage, the 20-foot reactor is divided by perforated plates into a number of separate fluidized beds operated is progressively changing temperatures.[18/] Because of the difficulties

18/ Bennett, B. B. The Gasification of Latrobe Valley Brown Coal. Presented at Conf. on Gasification of Lignite and Inferior Fuels, Belgrade, Yugoslavia, September 16-18, 1963.

encountered with excessive devolatilization in the coal delivery tube, only chars have been used until now for extended runs. Comparison of these results with those in the cocurrent apparatus cannot be made as yet because of the use of char and other differences in operating conditions. Preliminary results using this apparatus are shown in Table 3.

Table 3.-Hydrogasification of Latrobe Valley Coal

Volatile matter in char, percent		18.0
Pressure, psi		600
Temperature, °F.		1,380
Effective reactor length, feet	4	6½
CH_4 content outlet, percent	17.3	27.2
Char gasified, percent	50.2	63.3

SPECIAL GASIFICATION PROCESSES

Using entrained gasification systems with fine coal, high carbon conversions can only be attained with high oxygen requirements. The Rummel[19/] single-shaft

19/ Rummel, Roman. Gasification in a Slag Bath. Coke and Gas, Vol. 21, No. 247, December 1959, pp. 493-501, 520.

generator in which the oxygen-coal and steam are injected into a rotating slag bath is an apparatus designed to provide extra residence time to get more complete gasification at reduced gasifier volumes. Other potential advantages are its ability to use a high-ash coal, to operate with both coking and noncoking coals, and to use a larger sized feed. If air is used in place of oxygen, producer gas can be manufactured.

Another modification of the slag bath generator uses a double shaft instead of the single shaft, substitutes air for oxygen, and still produces a nitrogen-free mixture of carbon monoxide and hydrogen. The slag bath contained in the bottom of a circular shaft is divided into sections by two vertical dividing walls. On one side coal and steam are injected into the rotating slag bath, and the coal is devolatilized and partly gasified by the steam, using heat supplied by the molten slag. In the second section, the balance of the coal and some fresh fuel are burned to raise the temperature of the slag and to oxidize the slag constituents that were reduced in the gasification section.

An experimental program on the double-shaft generator is being carried out at the Bromly-by-Bow Works of the North Thames Gas Board to determine its feasibility and to predict the cost of making gas in a full scale plant. The plant was designed to have a capacity of 95,000 standard cubic feet per hour, and a gas composition of 3.9 percent CO_2, 45.2 percent CO, 50.5 percent H_2, and 0.4 percent H_2S (nitrogen-free basis) was expected.

In initial tests (June 1962) coal was fired into the combustion shaft with the separating curtains absent so that the slag could flow unimpeded from one chamber to another. The slag flowed easily at 2 feet per second with a mass flow of about 200 tons per hour. In subsequent tests coal was introduced into the gasification chamber but the seal between the two chambers was not used because even in its absence insufficient heat was transferred to the slag by the combustion of coal. In addition, the points of admission of the steam and coal in the gasification chamber were such that contact between the reactants was poor.

After suitable modifications, the heat transfer to slag was increased by raising the blast pressure from 18 to 100 inches of water gauge, and the slag circulation rate was increased to 500 tons per hour. Better contact between the coal and steam was obtained by admitting these reactants to the gasification chamber in a single tuyere submerged below the slag surface. After the seal between the two chambers was put into place, a gas output of 40,000 standard cubic feet per hour was obtained. Gas composition was:

	Percent
CO_2	8.5
CO	23.0
H_2	45.0
C_nH_m	0.3
CH_4	2.5
O_2	0.5
N_2	20.2

Overall efficiency was 20 percent (heating value of gas divided by the heating value of coal consumed), although the corresponding efficiency considering only process coal was 69 percent. Improvements in the tuyeres are being made that are expected to increase the efficiency to 30 to 35 percent.

An economic appraisal of the process is now underway and if it can be shown to be economically attractive, if 50-to 60-percent efficiency can be achieved, an attempt will be made to demonstrate that such an efficiency can be obtained by redesign of the gasifier.

SUMMARY

Although improvements have been made in many of the coal gasification processes that have been developed, the competition from new discoveries of natural gas and from oil gasification processes makes their widespread application difficult to attain both in the U.S. and abroad. Continued experimentation, however, is being carried out on the most promising processes, and additional reductions in the cost of manufacturing a suitable gas for distribution can be expected. While many of the foreign research programs in this field have direct application to U.S. conditions, because a lower heating value gas is being sought, there is some difference in emphasis between foreign and U.S. programs. Of special interest at present are the experimental projects on slagging fixed-bed pressure gasification and the direct hydrogasification of coal to a higher heating value gas.